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PLANT RADIOIODINE RELATIONSHIPS
A REVIEW

by
James C. McFarlane and Benjamin J. Mason
Radiological Research Program
Southwestern Radiological Health Laboratory

U. S. Department of Health, Education, and Welfare
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I. INTRODUCTION

The discovery of atomic fission brought with it many important health problems. One of these is the possible contamination of air, water, food, and forage with radioactive iodine. Since radioiodine may reach man in many ways, it is difficult to discuss all routes in one review. This report, which is primarily a review of the literature, up to January 1969, discusses only one intermediate in the passage of radioiodine from the source to man's food--the plant.

The two objectives of this report are:

1. To bring together in one document a summary of plant-iodine relationships.
2. To provide data to aid in the design of experiments to broaden our present understanding of the contamination of plants with radioiodine.

It seems logical to start the consideration of the iodine problem with a brief look at the possible sources of radioiodine. Quantities of radioiodine may be released to the environment in several ways. Among the most important of these are nuclear explosions (both atmospheric and cratering devices), nuclear reactor operations, and reactor accidents. Under certain conditions, especially in nuclear facilities, chronic contamination may exist. In 1959, the National Committee on Radiation Protection (NCRP)[84] suggested that 9×10^{-9} μCi/cc of air was the maximum permissible concentration (MPC) of ^{131}I allowable in a nuclear facility. Although this level is very important in connection with industrial operations, it is generally of no direct consequence to the public.

Most releases of radioiodine to the environment are of short duration lasting from a few minutes to a few days. Under these conditions one

critical pathway of radioiodine is the air-forage-cow-milk-man route.[20] In 1964, the Federal Radiation Council (FRC) [37] recognized the thyroid as the critical organ for the radionuclides of iodine. Since milk is one of the main vectors of radioidoine to the thyroid the FRC recommended that children be considered the critical segment of the population.

The concern with milk as a route of 131-iodine to man is based on the 70 kg (fresh weight) of green alfalfa or grass per day. Radioiodine is therefore important, not because of the amount deposited on the vegetation but rather because of the efficient passage of radioiodine through this food chain and its ultimate concentration in a child's thyroid.

Although milk is considered as the main source of contamination, other sources should be investigated. The following examples may best illustrate the relationship of contamination by milk versus contamination from leafy vegetables. During a period of atmospheric testing (1962), contamination in the milkshed of Salt Lake City was from 300 to 2000 pCi/liter. At the same time the concentration of leafy lettuce reached 2800 pCi/kg fresh weight[9] To receive the same amount of activity as that received from one liter of milk, consumption of approximately 700 grams of leafy lettuce (not head lettuce) would have been required. A large dinner salad contains approximately 100 grams of lettuce. Since it is more probable that a person would drink one liter of milk each day than eat seven large dinner salads, milk is clearly a more important source of radioiodine contamination. However, it is clear that green vegetables may become contaminated and, therefore, cannot be neglected as a contributing source to man's total radionuclide intake. Thompson[107] suggested that as much as 20 to 40 percent of the possible ^{131}I contamination may be attributed to products other than milk in some non-urban adult population.

The same principles of plant contamination still exist, whether plants are eaten by man or by a cow. There will certainly be differences in

the quantity of plant material consumed and the method of food preparation, but the physiological and morphological principles which control the uptake and retention of iodine by plants will apply in both food crops and forage plants. The overall objective in studying plant-iodine relationships is to allow predictions of possible human ingestion of ^{131}I. Two main questions are of concern in this report.

1. What is the rate of deposition?

   (To describe deposition, it is necessary to understand differences caused by species variation, environment, and the form of contamination. To evaluate this, plant morphological and physiological factors which control the rate of deposition on and movement into the various parts of plants must be understood.)

2. How long does the contamination remain?

   (Variations caused by the chemical and physical states of fallout and also the effect of various environmental parameters such as wind, rain, temperature, and humidity on the loss of iodine from plants must be evaluated. Differences caused by plant morphology and physiological parameters such as foliar absorption and translocation must be considered in order to completely evaluate the radioiodine-plant relationship. To be able to predict the transfer rates of iodine to cows, it is necessary to have some insight as to the chemcial changes in the form of the iodine which occurs in or on plant surfaces.)

II. DEPOSITION

The kinetics of iodine deposition on plants was first studied by Chamberlain and Chadwick in 1953[20]. In their research they recognized the need to express the amount of plant contamination in relationship to the radioactive cloud. The term they defined (V_g) was simply a ratio between the amount of activity deposited on a horizontal surface per unit of time to the amount of activity in a volume of contaminating air.

$$V_g \text{ (cm/sec)} = \frac{\text{deposited activity/cm}^2\cdot\text{sec}}{\text{activity/cm}^3 \text{ of air}}$$

The area of deposition in this equation is considered to be that area of ground which may be completely or partially covered by vegetation. The resulting units were the same as velocity (distance/time); therefore, the term was originally called "velocity of deposition." Since that time, the term deposition velocity has been used by many investigators to describe the contamination of plants by iodine. This generalized equation is, however, useful only to the extent that the parameters which control the transfer of iodine from air to a surface are understood.

The transfer of radioactive fallout from the air to the surface of plants and soils is a complex phenomenon. Although this phenomenon is not well understood, it can best be studied by dividing it into two fields on interest. The first is the study of the contaminant from the time it is produced until it reaches the location of deposition. This area of study involves the fields of aerosol physics, meteorology, and microclimatology. The second area of interest is the study of those plant factors which have a modifying effect on the environment and thereby influence the rate of deposition. An investigation of these factors involves the study of plant morphology and physiology.

A. Physics and Chemistry of Iodine Fallout.

To understand iodine contamination of plants, it is important to first understand something about the manner in which iodine is released and how it reacts in the atmosphere. All methods of iodine release to the atmosphere involve high temperatures (in nuclear detonations) or powerful solvents (in reactor fuel reprocessing) and result in the evolution of isotopes of iodine as gases[43]. Although the fission process produces some 131-iodine directly, the majority of the 131-iodine is derived from the precursors, 131-antimony and 131-tellurium[7]. When fission products are released to the atmosphere, the ingrowth of 131-iodine into the cloud is rapid because the two precursors mentioned above have half-lives of only 23- and 25- minutes respectively[7]. Other isotopes of iodine are also produced in the fission chain[30]. An inspection of Table 1 shows that ^{132}I through ^{135}I are produced in greater quantities than ^{131}I. A look at the half-lives quickly shows why they are generally thought to be of secondary importance in the contamination of milk. Iodine-133 with a half-life of nearly one day, is present in such large amounts that it cannot be overlooked as a possible health hazard. However, most attention has been given to ^{131}I since its half-life is longer and, after a short time, it is the main isotope of iodine found in fallout.

Table 1. Important Radioisotopes of Iodine Produced by Fission.*

Nuclide	Half-life	Fission Yield (%)
^{131}I	8.05 days	2.9
^{132}I	2.26 hours	4.4
^{133}I	20.9 hours	6.5
^{134}I	54 minutes	8.0
^{135}I	6.75 hours	6.3

*Adapted from Bolles and Ballou, 1956;(7) Radiological Health Handbook, 1960 (30).

When a nuclear cratering device is exploded, a cloud is created which includes fission products, activation products, and part of the inert material surrounding the detonation site. Particulate fallout is largely the result of the attachment of radioactive nuclides to small particles of the inert or carrier material. The attachment of any particular nuclide is determined by physical and chemical properties of both the particle and the contaminant. Shleien[99] suggested that attachment of nuclides to particles of different sizes may vary with respect to the half-lives of their parents. To visualize this it is necessary to consider two processes.

1. Size fractionation (the size distribution of fallout particles diminishes as the time or distance from the detonation increases).
2. Radioactive decay (nuclides with short-lived precursors have a faster ingrowth rate than those having long-lived precursors).

These factors combined with the differences in the half-life of the various radioisotopes of iodine (see Table 1) had an effect upon the ratios of ^{133}I to ^{131}I in the fallout from a test conducted at the Nevada Test Site.

In April 1966, the Atomic Energy Commission's (AEC) Project Pin Stripe accidentally released a small amount of contamination from an underground test. An experiment (unpublished) conducted by this laboratory showed that the ratio of ^{133}I to ^{131}I deposited on plants decreased as the distance from the hot line increased. Since the half-lives of the precursors of ^{133}I are much shorter than those of ^{131}I, it is understandable that the former was more plentiful in the region close to the cloud center.

Radioactive fallout consists of a complex mixture of radioactive gases and various sized particles. The meteorological parameters that control mixing, movement, and dispersion of a fallout-cloud determine when and to what rate portions of the cloud reach the surface of the earth. At the surface, micrometeorological factors control the deposition of the radioactive contamination. The physics of the actual removal and retention process of small particles and gases is complex and therefore not well understood.

In 1966, Fisher[38] reviewed the subjects of deposition of iodine vapors and contaminated aerosols on plants. He suggested that the aerodynamic factors which control deposition of both aerosols and vapors at the air-ground interface are gravity, electrical forces, thermal forces, Brownian motion, molecular diffusion, and turbulence. Describing the deposition of particles, Chamberlain[17] considered the primary transport mechanism in the turbulent boundary layer to be eddy diffusion with molecular diffusion being effective over the last few millimeters. Outside the boundary layer the particles are subject to the well known Stokes' relation for terminal velocity and as a result possess a constant downward velocity. Considering the various parameters, Fisher[38] developed a model for the deposition of vapors and aerosols on leaves. Using this model, he compared some of the reported values to the theoretical predictions. He found that he was only accurate to within a factor of two for aerosols and a factor of three for vapors. His model holds that increased wind speed causes a corresponding increase in the deposition velocity and that an increase in particle size from 0 to 20 μm causes an increase in deposition velocity from 0.25 to 3.0.

The difference between the observed deposition velocity and that predicted by Fisher may possibly be explained by understanding more about the morphology and physiology of the plant surface. The velocity of deposition and retention of particles on leaf surfaces is largely dependent on the thickness of the boundary layer of air at the surface of the plant[3,17,22]. The thickness of this boundary layer is influenced both by physical factors and plant factors. Where the boundary layer is thin, contaminants are deposited in greater abundance. This could explain why iodine is often concentrated at the margins of leaves[20,54]. High velocity wind reduces the boundary layer to a very thin film next to the leaf surface. In contrast, low velocity winds allow the boundary layer to thicken. The morphology of the leaf surface also determines the shape and extent of the boundary layer. Epidermal hairs act the same as a windbreak in reducing wind speed close to the surface. This reduction in speed causes the boundary layer to thicken, i.e., a leaf which is covered with epidermal

hairs has a thicker boundary layer than a smooth leaf. Thus, by modifying the boundary layer, variations in morphology can be responsible for variations in retention of airborne nuclides. The physiology of the plant effects the boundary layer by changing the rate of gaseous exchange, by changing the relative humidity adjacent to the leaf surface, and by positioning the leaf in different attitudes with relationship to the environment.

The rate of settling the deposition of iodine depends on the physical form of the contamination[34]. One of the limiting factors in our understanding of the physical form of the deposition is the difficulty incurred in sampling the atmosphere in a meaningful way.

Particulate contaminants are generally classified by one of three methods: graded filter,casade impactor, or photographic methods. Two types of filter systems are used for sizing particles. One type uses filters with several different pore sizes placed in series. Microfilters are very efficient for this type of determination since they are essentially 100 percent efficient for particles larger than their pore size. They are available in ten porosity grades from 0.01 to 5 μm[63]. Another system uses only one type filter and measures the depth to which various sized particles penetrate into this filter. Silverman[100] used a polyester filter to collect and classify particles from 0.002 to 0.35 μm diameter. One limitation of both of these systems is the fact that the filter characteristics change with time because of the particles which are collected on or in the filter material. A particle may only partially cover a pore; therefore, limiting the size of the particle which will pass through that pore. Especially when the air has a high dust load, the efficiency of the filter changes and thus particle sizing becomes less accurate as the operation progresses.

Inertial particle or cascade collectors make use of the fact that particles moving in an airstream tend to follow along their original direction when the airstream is deflected by an obstacle. Impactors collect the particle on the surface of the deflector. Impingers use

the same principle but collect the particle in a liquid. By having a series of impactors with a different airspeed at each step in the sampler, particles of different sizes will be collected. Cascade impactors are generally not efficient for particles less than 1 μm.

The final method of measuring fallout particles is via microscopic measurement. Particles for these measurements are often collected on planar surfaces and sized visually by the aid of a photograph taken through the use of either a light or an electron microscope. Although this is a very time-consuming and laborious task, there is one advantage in this method over the previous two. In deposition studies we are interested in the distribution of deposited or settled particles rather than the total in the air sample. Since a planar collector resembles the surface of a plant more than a high-volume air sample, the ratio of various-sized particles on such a plate would be more representative of the distribution on a plant surface. (A plant is not, however, simply a planar surface, therefore, some difficulties arise in interpreting this type of information.)

Detection of iodine gas is equally as difficult as detection of the particulate materials and there is, perhaps, even more possibility of error in this determination. The most commonly used method of classifying fallout into its two major fractions (particulate vs gas) is done by collecting the particles on an inert prefilter and the gases, which pass the prefilter, on activated charcoal.

Since activated charcoal is nearly 100 percent efficient for the collection of most of the gaseous iodines, the resolution of this system is largely dependent upon the efficiency of the prefilter for removing the particulate material from the air sampled. Small particles are perhaps the biggest source of error in this system. To have a system which collects all submicron-sized particles on the prefilter generally requires some type of graded filter system. Without this there is a good possibility that much of the activity seen on the charcoal filter is in reality very small particles rather than gaseous iodine.

Other possible sources of error are related to the adsorption of gaseous iodine onto the prefilter and also to the possible revolatilization of

some of the iodine from the charcoal during the period of time between sampling and the time the filter is collected.

Kuhn[63] gives information on methods for analyzing air for very small particles.

Keeping the above limitations of collection systems in mind, the following material will examine the data in which particulate and gaseous iodine has been investigated.

Eggleton, et al.,[33] classified world-wide fallout from the Russian atmospheric testing of 1961. This study, which used filters to size the fallout, covered a three-month period following the tests. As a prefilter they used a high-efficiency asbestos filter followed by brass gauze for elemental iodine and charcoal-impregnated filter paper for removal of certain other compounds of iodine. This filter system was backed by a one-inch bed of activated charcoal. These authors found that an average of 75 percent of the ^{131}I contamination was in particulate form.

Megaw[74] contaminated the inside of a reactor shell with radioiodine in order to study simulated conditions of a reactor rupture. He found that from 40 to 80 percent of the iodine released had become attached to particulate material within the first hour. A large amount had also become attached to the containment vessel walls. Much of the particulate iodine can be accounted for by very small particles called Aitken nuclei[16]. These are particles of about 10 nanometers in diameter which are formed in all combustion processes and are therefore generally present in air. Aitken nuclei are so small that they are sometimes thought to move in a manner similar to gases. Even though this may be partly true, there are some differences which should be recognized.

Under the influence of gravity the Aitken nuclei has a settling velocity which is different from that of a gas. The important difference is the fact that the Aitken nuclei consolidate to form larger particles as they are carried from the point of release. These particles and their aggregates therefore do not respond to changes in meteorological conditions in the same manner as a gas.

The studies of Perkins[90] have shown that a large and varying fraction (from 10 to 90 percent) of the radioiodine in fallout is in the gaseous form. He tried to characterize the chemical form of the gaseous iodine and found that very little, if any, exists in the elemental or HI form. However, the gaseous compounds of ^{131}I in air were not identified. Up to 20 miles, there was an increase in the amount of particulate iodine contamination in a cloud with increased distance from the release (Figure 1).

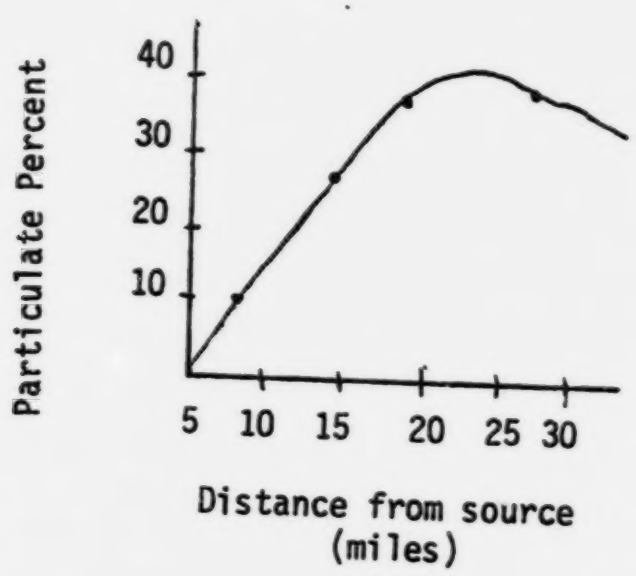

Figure 1. Physical Form of ^{131}I in Air at Various Distances from the Source[90].

Nishita[84] has stated:

> "The chemical and physical properties of fallout depend on the energy yield of the nuclear device, the degree of intersection of the fireball with the ground surface, the mineral composition of the ground surface, and the structural material surrounding the device. Nuclear devices detonated on or near the surface of the ground have been found to yield predominantly siliceous fallout particles because of the incorporation of soil into the fireball. Particles from detonations at higher elevation more nearly reflect the incorporation of the structural materials surrounding the device. A large fraction (>50%) of the close-in fallout from nuclear devices detonated on steel towers at the Nevada Test Site was attracted to magnets and was red-brown in color suggesting the formation of magnetite. By comparison, the devices that were not detonated on steel towers produced fewer magnetic particles (<10%). Devices

> fired at the Pacific Proving Ground produced fluffy conglomerates of $CaCO_3$ from coral and crystalline NaCl particles from sea water. Thus, the chemical composition of fallout may vary considerably depending upon the conditions of detonation."

It seems obvious that the ratio of gaseous to particulate contamination and the size distribution of particles is dependent on the amount of particulate material in the cloud and the distance or time from release. Shleien[99] found that airborne fresh fission material occurred on larger particles and that the size of the contaminated particles decreased with time. Most of the older fission products were associated with particles having diameters less than about 1.75 μm.

B. Humidity.

Humidity has been recognized as a factor which influences deposition of iodine[34]. Cline and Hungate[25] observed that moist leaves of a plant species accumulated up to 2.2 times as much $^{131}I_2$ as did dry leaves of equal area. The same was true with moist paper but there was only a very slight increase in accumulation noted for moist soil. Barry and Chamberlain[3] offered two possible explanations to account for these observations. "It may be that humidity was responsible for regulating the size of the stomatal aperture. Alternately, adsorption of iodine on the external surface of the leaf may have been in some way facilitated by conditions of high humidity."

Let us consider transpiration, not because of any suspected relationship with iodine diffusion, but rather because it might give us some information on which to form a hypothesis. The rate of transpiration through open stomates is regulated by the gradient between the vapor pressures of water inside and outside of a leaf. In the substomatal cavity the humidity remains very close to saturation. The vapor pressure gradient is therefore determined by the temperature of both leaf and air and the relative humidity of the air. When the gradient decreases, the rate of transpiration also decreases until at 100 percent relative humidity (RH) transpiration stops.

One would intuitively think that transpiration would also be greatly influenced by the degree of stomate opening. This is, however, not the case. Diffusion through the stomate is not correlated to the area of the individual stomate but to the circumference of the opening[62]. This means that small changes in the amount of opening and closing are of little importance in restricting diffusion.

With this in mind, let us consider how humidity might alter the diffusion rate. Atmospheric humidity does not directly affect the degree of stomatal opening. Under high atmospheric humidity the gradient of pressure potentials between the atmosphere and the substomatal cavity is decreased and the rate of transpiration would therefore be suppressed. This would cause a decrease in the rate of water loss and could, under conditions of water deficits, cause a decrease in plant water stress. Under such conditions greater tugor would result in the guard cells and the stomatal aperture would open larger. This effect would only occur under special conditions and would not be a general response to increased atmospheric humidity. In addition to this, it is not logical to presume that humidity may change the concentration of iodine in the air. A more probable explanation would be related to the chemical properties of iodine in a humid environment and the physics of deposition and retention of particles and vapors on a moist surface.

The importance of humidity in the deposition of iodine on plant surfaces is still undefined. There is no data available which would allow a statistical evaluation of the effect of various amounts of atmospheric moisture upon deposition. The effect of humidity on retention and absorption of foliar-applied iodine is likewise not understood. Since humidity has been documented as one of the important factors in determining the rate of contamination, attention should be give to quantitating this effect.

C. Species.

The basis of plant classification is the anatomy of the flower. There are other differences, however, which are of greater consequence in the contamination of plants by radioiodine. Each species of plant can also be characterized by leaf differences such as size, shape, surface, number of leaves, and their orientation on the stem. Plant leaves are special organs which function mainly to absorp radiant energy and exchange gases with the environment. Both of these functions require leaves to present a considerable surface to the atmosphere. Herein lies the key to the importance of plants in the passage of iodine to man. Because of their large area and ability to exchange gases, leaves are efficient collectors of radioactive fallout. Let us therefore consider some of the differences in leaves as they relate to the collection of radioiodine

1. Plant growth habit.

The fact that species differ in their collection ability for radionuclides is exemplified by the data of Gorham[47] and Davis[29]. These authors, working with ^{90}Sr from fallout, showed that mosses and lichens accumulate far more of this radionuclide than do vascular plants. This difference was considered to result from the differences in the growth habit of these plants. Similar results can be expected with other radionuclides.

Preliminary data obtained at this laboratory indicate that 131-iodine from fallout is retained differently by different species of plants. A dense stand of alfalfa retains more iodine than a more open stand of sudan grass. This again is considered to be a result of both growth habit and leaf morphology.

Natural desert vegetation such as sagebrush tends to have an open (less leaf area per unit of space occupied by the plant) character, thus the air is free to move through the plant and expose more of the leaf surface area to the contaminant than would be encountered in a dense plant such as alfalfa.

An alfalfa field has only the top few inches of the plants situated in an area of great air movement. The lower leaves are mostly protected from wind and air movement. Figure 2 shows the profile of air movement in a stand of alfalfa growing on a research farm managed by the Southwestern Radiological Health Laboratory for the U. S. Atomic Energy Commission.[122] It is obvious that the lower leaves are in a different environment, therefore, have a different exposure to contamination than the upper leaves.

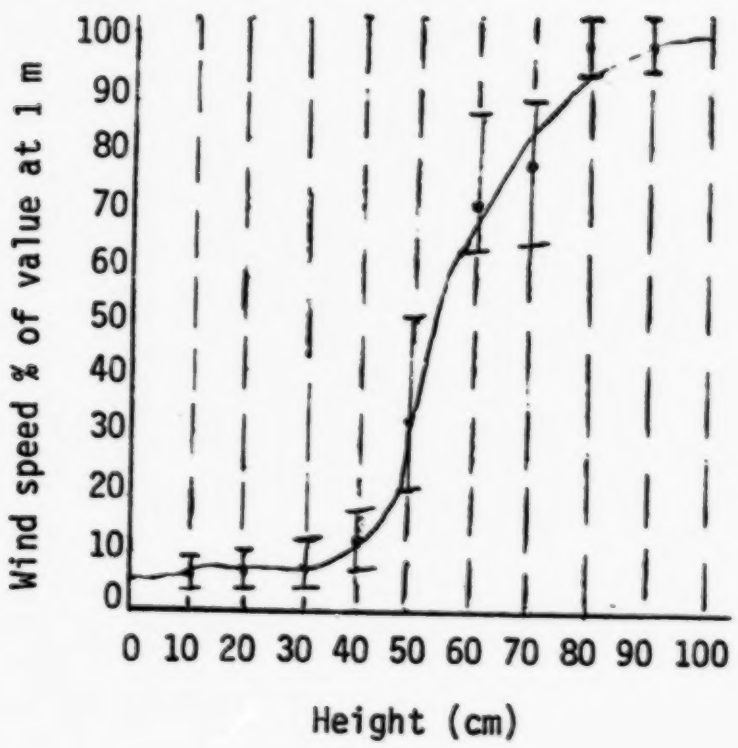

Figure 2. Air Movement in Alfalfa Field.[122]

Cline, Wilson, and Hungate[27] observed that rye plants collected gaseous iodine mostly in the middle portion of the plant. The top collected 39 percent, the middle 49 percent, and the bottom 18 percent. They also observed that this resulted from the seed heads causing the tops of the plants to hang down so that the middle of the stem was actually the highest and most exposed portion. Barry and Chamberlain[3] observed that the smaller leaves at the top of the stem were often associated with higher absorption than the lower leaves. Bunch[13] reported greater absorption of iodine on the upper parts of grass leaves. From the above observations, it seems apparent that the amount of contact with the contaminated air appears to be greatest at the top of a plant and that this is the area of greatest contamination.

An important point to remember in evaluating contamination of plants by radioactive fallout (particularly iodine) is the method of expression. Since the leaf is the plant part in greatest contact with the environment, it is the plant organ on which deposition is ultimately dependent. The number, size, and type of leaves presented to a radioactive cloud are therefore the most important plant factors in determining the rate of accumulation or deposition. Under laboratory conditions, Bunch[13] showed that the deposition velocity rises logarithmically with an increase in the density of vegetation. Gifford[46] showed that vegetation, specifically sagebrush and grass, collected more contamination per area of ground cover than either bare soil or flat plate collectors. Cline[27] observed the deposition velocity on bare soil and found it to be approximately half that found on living plants. Plant cover presents a larger collection area than a bare soil or a planar fallout collector. This difference could account for the difference in observed deposition velocities between plants and soils. By reexamining the equation commonly used for deposition velocity, we find that it is based upon the ground surface area. The observations cited above make it obvious that the rate of deposition on plants should be based in some way upon the effective collection area of the particular plant involved. The leaf area of a plant is a

difficult parameter to obtain. The effective collection area (includes all epidermal surfaces of the leaf and stem, all protrusions, the effective gas exchange area created by the stomates, and all surfaces of bark and other dry material on the plant) is even more difficult to measure than leaf area. There must be, between the ultimate and the obvious, some parameter which will allow us to gain a better understanding of the deposition of radioactivity upon plants.

Some authors have based contamination data on the fresh weight of the sample. It has been found in this laboratory that the fresh weight of forage samples can vary as much as ten percent due to the time between the collection and the weighing of the sample. Placing the samples in sealed plastic bags can certainly reduce the loss of moisture but cannot eliminate it.

Other sources of variation in the moisture content of vegetation are the time since the last irrigation or rainfall and the relative humidity of the air. Figure 3 shows the change in the percent moisture of an alfalfa field between periods of irrigation. If the loss of contamination from alfalfa were studied using for a basis of expression the fresh weight of alfalfa, an erroneous interpretation of the data could easily result. Also comparison between species can be confounded by differences in their moisture percentages. Table 2 shows the variation of the moisture content of four species collected at the same site at the same time.

In order to overcome the above mentioned difficulties resulting from the use of fresh weight measurements or those resulting from the use of ground surface area, some authors have used the sample dry weight as the basis for expressing vegetation contamination data. This method has the advantages of 1) reflecting the amount of material and 2) avoiding the problems of moisture variation associated with fresh weight methods.

The method used by this laboratory[122] is to keep the sample in a sealed plastic bag from collection until weighing. If the sample cannot be weighed immediately it is kept in an ice chest or refrigerator until

Table 2. Variation in the Percent Water of Four Plant Species on Three Different Collections.(122)

Date	Percent Water			
	Arar*	Epne†	Orhy*	Sihy†
27 March	55.3 ± 4.1	48.5 ± 5.0	16.0 ± 2.6	59.0 ± 5.2
22 April	62.0 ± 2.0	49.7 ± 1.5	41.7 ± 7.6	63.3 ± 4.6
4 June	53.0 ± 2.0	44.3 ± 1.1	51.7 ± 2.1	54.6 ± 1.6

* = *Artemisia arbuscula* (black sagebrush)
† = *Ephedra nevadensis* (Mormon tea)
* = *Orhyzopsis hymenoides* (Indian rice grass)
‡ = *Sitanion hystrix* (squirrel tail grass)

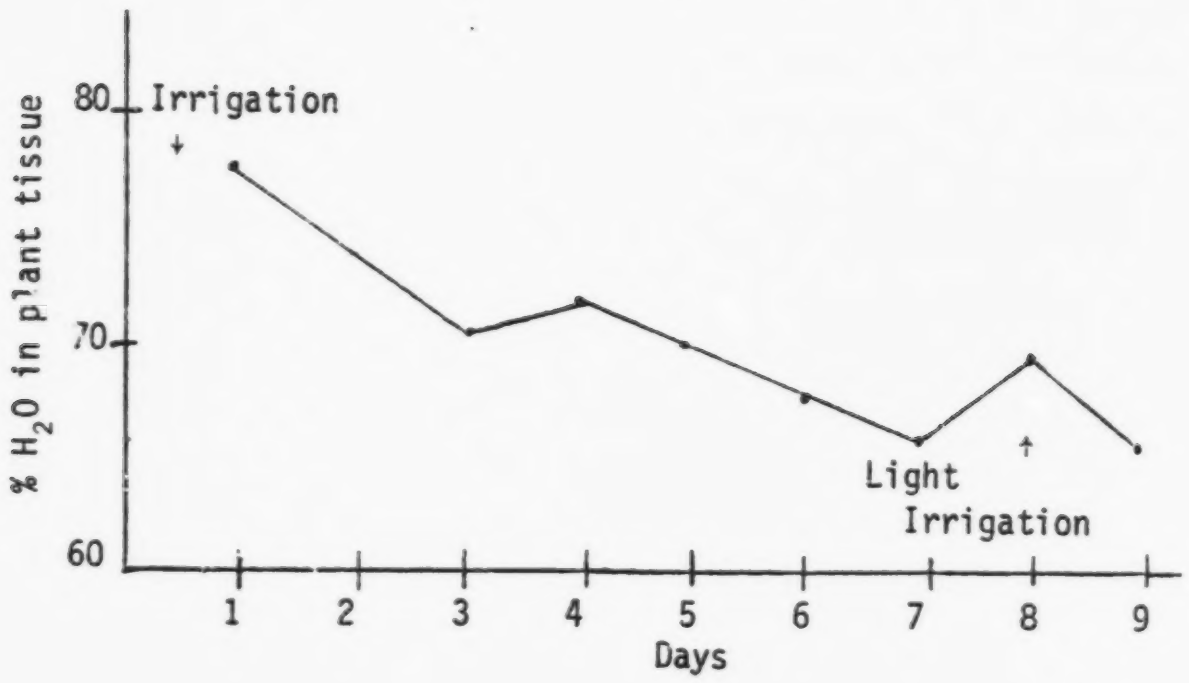

Figure 3. Percent Water in an Alfalfa Stand Between Irrigations.(122)

Note.--The period between irrigations was about twice as long as the general practice. This was done in order to prevent interference with an experiment. Notice that the crop did not recover to the original percent water, presumably because of the extended drought between irrigations.

the time of weighing. After weighing, the samples are counted then returned to the principal investigator who is responsible for drying the sample to a constant weight and reweighing the sample. (It has been found that drying at 75°C for 24 hours or longer will provide this condition.)

The two main disadvantages of the dry weight method are the increased costs due to extra handling and failure of this method to account for the dilution of the activity resulting from plant growth during the time of an experiment or study.

At this time, basing data on a dry weight basis appears to be the best available method. The ideal however would be to have a rapid means of measuring the actual surface area of the plant that is exposed to the contamination. Efforts are underway in this laboratory to try to develop a suitable method to determine such a measurement.

Also, it must be remembered that plants change in response to climatic variation; therefore, the crown density, leaf morphology, and physiology will not always remain constant. Crown density, for instance, may be much different on plants of the same species at different locations, at different times of the year, or at different periods of development. Leaf morphology may change at different sites or different seasons; i.e., some leaves shed epidermal hairs or curl in response to drought. Likewise in an area of ample moisture, leaves are generally larger and more succulent than in arid regions. Time of day, wind, and available water cause changes in plant physiological functions. Transpiration, gaseous exchange, and leaf positioning are changed in response to the environment. Because of these variables, contamination of plants will always be a complex subject that will only be understood as each factor is evaluated.

2. Leaf morphology.

The purpose of this paper is not to review leaf morphology but simply make it clear that there are many differences in leaves, some of which may be very important in the collection and retention of radioiodine from fallout. A review of the anatomy and morphology of leaves can be found in "Plant Anatomy" by Esau[36]. There has been very little information reported concerning the effects of different leaf types in relationship to iodine contamination. In 1963, Hungate, et al.,[54] reported on an experiment where plants of different leaf characteristics had been contaminated by the effluent gases of a simulated reactor disaster. Two of the species used in this field test were of extreme difference in leaf morphology. The hairy leaves of geranium had no more iodine contamination per unit area of leaf surface than the smooth leaves of *Peperomia*. Contrary to this, Romney[94] reported the results of Project Teapot where he observed that hairy plants accumulated more fallout than smooth plants.

Creosote bush, *Larrea divaricata*, provides an example of another plant characteristic that effects the retention of radionuclides. The leaves of this species are covered with an exudate which gives the leaf a sticky surface. This material acts as a trap for much of the particulate contamination which makes contact with the leaf. A comparison made in this laboratory revealed that the leaves of this plant are as sensitive a particle collector as are planchets which have been covered with an alkyd resin.

The use of plants as an indicator of the presence of radioactive contamination has an advantage over planchets in that it is not necessary to pre-place the collector in the expected path of the contamination cloud. Due to the stickiness of the leaves, the Southwestern Radiological Health Laboratory utilizes creosote bush, when available, to determine the location of the deposition

"hot line" which results from those tests conducted on the United States Atomic Energy Commission's Nevada Test Site which release radiation to the environment around the test area. Samples of vegetation are collected and counted for gamma activity.

Although some plant species have been regarded as better collectors of fallout than others and some authors[54, 94] have reported differences between species (casually observed) in their ability to retain radioactivity, the effect of leaf morphology upon collection or retention of fallout has not been evaluated conclusively. This is an area of needed investigation.

III. ABSORPTION OF IODINE BY PLANTS

A. Roots

Independent of the advent of nuclear weapons and the threat of fallout contamination, iodine in plants has been observed and reported (Orr, et al.,[86], 1948; Bohn[6], 1917; Campbell and Young[15], 1949; South Carolina Agricultural Experiment Station[103], 1929; Chilean Iodine Educational Bureau, Inc.[24], 1948, [23]1960; Vogel[115], 1934; and Malhotra[66], 1931). Selders[97], 1954, was the first to report the effect of different substrate conditions on the uptake of iodine. He experimented with plants both in soil and in hydroponic cultures and came to the following general conclusions:

1. The percent iodine in the plant tissue responded directly to the levels in the substrates. At a level of six micrograms of iodine (as KI) per gram of soil the plant concentrations reached a maximum of aprroximately twelve times that found in the substrate.

2. Iodine uptake increased by a factor of four when the pH was changed from seven to four. This was probably due to the effect of the hydrogen ion upon the cell membrane rather than any change in solubility or availability of iodine.

3. The four species tested absorbed iodine in different amounts. The order was bean>tomato>barley>Russian thistle.

4. Iodine was only slightly translocated from the site of original deposition. Some iodine was lost from the roots, apparently, being translocated to the primary leaves rather than to the culture solution.

Iodine is regarded as a physiologically non-essential element for most plants. It is possible that iodine may partially substitute for chlorine in some plant functions. However, at concentrations

above 1 µg/ml it produces toxic symptoms in plants and therefore interfers with observations[10]. Uhler[113] found that below toxic levels iodine absorption and translocation were independent of both photoperiod and transpiration. In 1965, Uhler[114] reported on relationships of iodine uptake and different metabolic inhibitors. Iodine uptake was found to be independent of transpiration and followed a different pathway than cations under the same conditions. Uptake was also temperature dependent and was decreased by metabolic inhibitors. This evidence suggests that iodine absorption by roots is dependent on a source of energy and is therefore an active uptake process.

The point of maximum uptake of iodine in roots appears to be within a few millimeters of the root apex and is not enhanced by the presence of root hairs[44]. In the studies reported in this section, iodine was present in the substrate in the iodide form. Once inside of the plant, the majority of iodine remained in the iodide state. Of the organic compounds of iodine which have been observed in plant tissues, three have been identified as amino acids. They are 3:5-di-iodotyrosine, 3:5:3-tri-iodothyronine and 3:5-di-iodothyronine[40].

Iodine contamination is not considered a soil or root problem because of its short half-life. However, it is soluble in many forms, is able to percolate into the soil, and (as discussed above) is concentrated by plants when in the root substrate. The limiting factor in this route of plant contamination is the reactivity of iodine with the organic and clay components of the soil[68]. Iodine is mostly held in the top few centimeters of the soil even against large amounts of leaching water. By the time radioiodine can reach the root zone, be absorbed into plants, and translocated to the leaves, it is of little consequence as a radioactive element.

The portion of iodine which reaches the edible portion of forage plants has had time to decay and much of the radioactivity is lost. Soils contamination therefore is not considered to be a serious problem in the transport of radioiodine to man.

B. Leaves

Iodine from fallout has been observed to be absorbed into leaves. The extent of this absorption in comparison to that simply adsorbed to the surface is important because of the possibility of decontaminating the plants by removing the latter in some cultural operation such as irrigating. Hungate[54,55] and Selders[96] followed the penetration of iodine from three different sources into leaf mesophyll. They found that when leaves were exposed to elemental ^{131}I, 35 to 40 percent penetrated to the mesophyll. When leaves were dipped in a solution of $Na^{131}I$ no penetration was observed. Plants contaminated by the effluent from a simulated reactor accident had only 10 percent of the ^{131}I in the mesophyll of the leaf.

One would expect that iodine in the form of a gas could enter plants via the stomates. Meyer, et al.,[76] listed the size of fully opened stomates of 14 different plant species. Assuming an elliptical shape, the calculated area of the stomate opening ranges from 17 to 294 square micrometers. The size of gas molecules is assumed to be considerably smaller than this because gaseous water molecules, which are relatively complex, are known to readily pass through the stomate opening. Aitken nuclei and even larger particles up to perhaps 0.5 micrometer in diameter are also small enough to enter the stomate opening.

IV. HALF-LIFE OF IODINE ON VEGETATION

The motivating force behind most investigations of radioiodine has been the desire to minimize the hazard of this pollutant (one of the most prevalent in fresh fission fallout). The degree of plant contamination at the time of ingestion by cows is the primary factor determining the amount of radioiodine which appears in milk. The amount of contamination a cow receives is determined by the amount of deposition minus the amount lost before consumption. An understanding of the loss of iodine from plants is therefore equally as important as an understanding of its deposition. Iodine-131 decays to ^{131}Xe (stable) by both β and γ emission with a radioactive half-life of 8.05 days. Activity is also lost by other processes. The effective half-life (T_{eff}) on plants is therefore defined as the cumulative effect of both radioactive decay and all other loss processes. Reported values of T_{eff} (Appendix B) range from 3.5 to 6.5 days. Plants may lose contamination by three methods--dilution by plant growth, loss of physiologically incorporated iodine, and physical loss of surface-attached iodine. For a review of effective half-life of iodine on plants refer to Thompson[108] and Chamberlain and Chadwick[21].

A. Growth

Loss of contamination by growth is rather straightforward. A given amount of contamination is simply diluted as plants increase in size and weight. If contamination were expressed on the basis of ground covered by vegetation, this effect of dilution would be masked. In some respects this masking may be desirable, but since a cow is interested in a quantity of feed and not on the area required to produce the feed, it seems only logical that data expression must be on a weight basis. The effect of plant growth is obviously most important during periods of rapid growth.

As an example, consider an alfalfa field one week before cutting. Under good conditions, an alfalfa field may produce 1½ to 2 tons per acre in a five-week period. If the field were contaminated by 131-iodine and cut and fed one week later, the contamination would be decreased by 45 percent due to radioactive decay and another 20 percent due to dilution by plant growth. It is obvious that under some curcumstances this type of loss may be of considerable importance.

B. Absorbed Iodine Loss

Translocation of iodine in plants away from the site of absorption has been studied by Fowden,[40] Hungate et al.,[54] and Selders and Rediske,[97] and found to be very slow. Experiments in this laboratory have shown that after 72 hours in hydroponic cultures containing $Na^{131}I$ the distribution of ^{131}I in bean plants is as shown in Table 3.

Table 3. Distribution of ^{131}I in Bean Plants Growing in $Na^{131}I$ Contaminated Hoagland's Solution.

Root	96.5%*
Stem	2.1%
Leaf	1.1%
Fruit	0.2%

*Percent based on dry weight of tissue.

Based on these observations we would conclude that the translocation of absorbed iodine is very slow and that it is only of minor importance in determining the effective half-life of iodine on plants.

C. Adsorbed Iodine Loss

Chamberlain and Chadwick[21] commented on five methods in which iodine may be lost from vegetation, four of which describe adsorbed iodine loss. Their conclusions are based on reported literature and are as follows:

1. Rain water is of secondary importance in washing iodine from plants.
2. Volatilization has been reported as both important and unimportant as a mechanism of iodine loss from plants. The importance of this is still uncertain.
3. Translocation to other parts of the plant is not rapid nor important as far as loss of iodine is concerned.
4. Dieback may cause some parts of the plant to escape sampling.
5. Plants may shed and regenerate parts of their cuticle.

There have been many differences in the reported half-lives of ^{131}I on vegetation (Appendix B). Some of these are undoubtedly a result of differences in the mode of expressing results. Other differences may be due to environmental variables and the chemical or physical form of the contamination. There is no clear definition of the relative importance of the environment upon the loss of the contamination. Different forms of iodine (particulate, gaseous, or liquid) seem to be attached to or absorbed in plants with varying degrees of affinity under different environmental conditions. Therefore, it seems logical that the loss of the iodine would also be dependent on different forces. Whether these forces act cumulatively or independently and what the major mechanism of loss is, remains a very complex and challenging problem. The full explanation for the relationship between radioactive decay and effective half-life is, at this time, not understood.

Straub[105] reported that when cows were taken off ^{131}I contaminated feed the effective decay occurred in two phases. The first portion had a half-life of 16 hours to two days. Later the effective half-life leveled off to about seven days. Experiments in this laboratory with contaminated dry aerosols indicate the same type of decay scheme in plants. Immediately following contamination, ^{131}I is lost from plants rapidly. The contamination is so loosely attached that it can be blown off by wind or washed off by rain or irrigation. After

a short period of time the contaminants become bound to the surface and incorporated into the plant in such a way that they are dislodged only very slightly by changes in the environment. A moderate amount of experimentation in this lab indicates that the species of plant and the physical and chemical form of the contaminant determines the rate of fixation as well as the resultant loss rate. We have observed that this first period lasts from one to four days and has a T_{eff} of from one to three days. After the end of the first phase, ^{131}I is lost from the plants much more slowly, i.e., T_{eff} ≈ five to seven days. Decay in this portion of the curve is more a result of plant growth and radioactive decay of ^{131}I than the loss of contaminant. Figure 4 shows the decay of gaseous elemental ^{131}I on alfalfa plants.

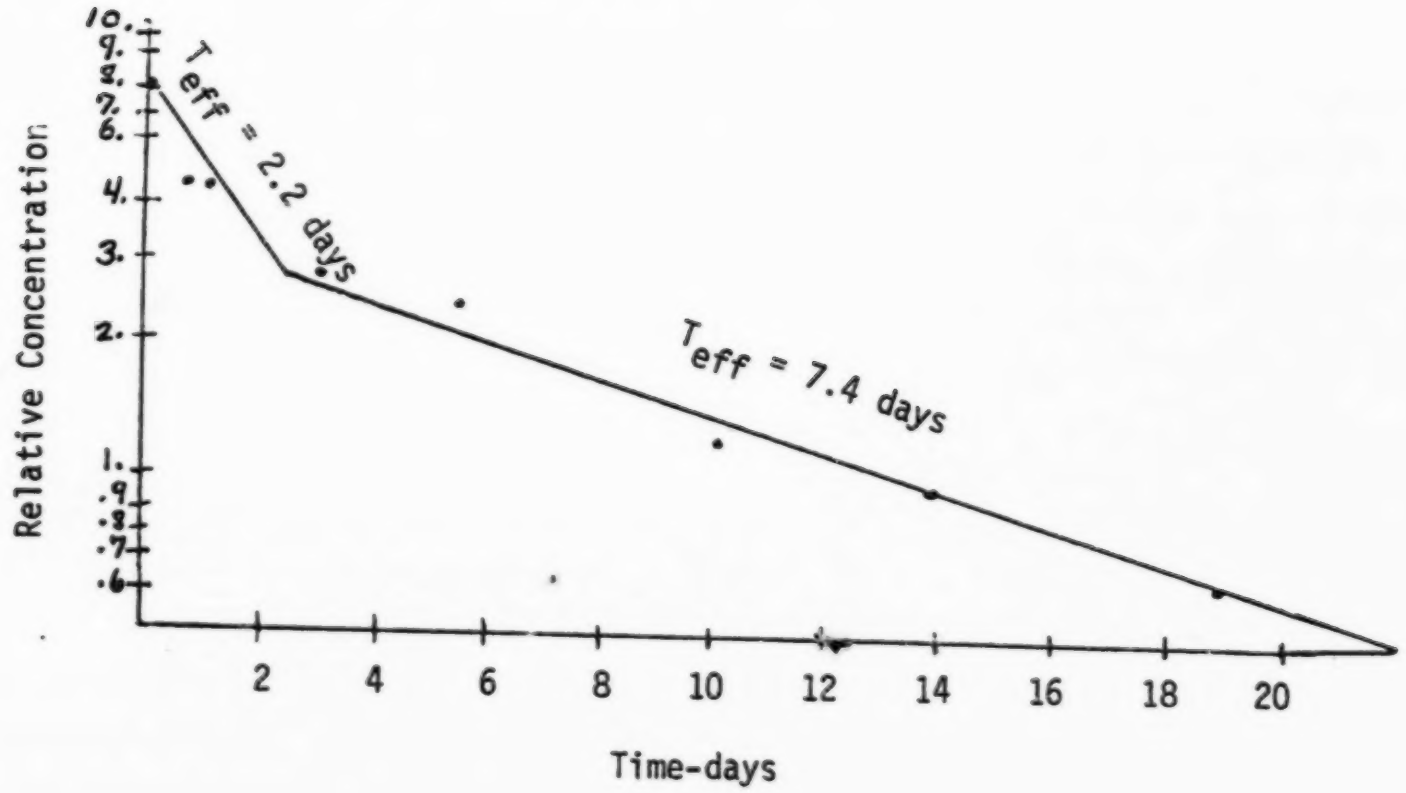

Figure 4. Decay of $^{131}I_2$ from Contaminated Alfalfa Plants.[122] (Each point is the mean of four observations.)

V. SUMMARY AND CONCLUSIONS

The term deposition velocity has been widely used to describe iodine contamination of plants. The physics of iodine transfer from air to plants is a complex problem and involves both chemistry and physics of the fallout, various forces which cause movement of the particles in the atmosphere, and the behavior and extent of the boundary layer which surrounds each leaf surface. In 1966, Fisher[38] developed a model which can be used to predict the deposition velocity to within a factor of three. Increases in wind speed or particle size cause a corresponding increase in deposition velocity. It has been recognized that different plant species collect contamination at different rates. These differences have not been clearly defined, but before a complete understanding of deposition velocity can be attained, differences in deposition on various species must be understood in relationship to their physiology and leaf morphology. Some of the factors which should be evaluated are differences in deposition under light and dark conditions, differences caused by variations in leaf morphology, and differences caused by change in humidity.

Iodine is considered as a non-essential element for plant growth. Absorption of iodine through the roots has been shown to be dependent on the iodine concentration, pH, and plant species. Although it has not been documented, it appears that iodine absorption is by an active uptake mechanism. At levels above 1 μg/ml in a culture solution, iodine was found to be toxic to plants. After being absorbed by a plant tissue, iodine is only very slowly translocated. Contamination of plants through the roots by fallout iodine has been shown to be of little importance in the total contamination of plants. Foliar absorption, on the other hand, provides a route whereby iodine can be fixed against most decontamination measures. Foliar absorption of iodine by plants is largely dependent on the chemical and physical form of the contaminant. Very small particles and gases can enter

the plant through the stomata. Dissolved iodine enters through the cuticle mainly in the areas of ectodesmata. The rate of iodine absorption by plants from different forms of contaminant is a field which warrants added investigation.

The effective half-life of iodine on plant tissues includes both radioactive decay and other loss processes. Effective decay rates have been reported from 3.5 to 6.3 days. There are three methods of loss which need to be considered in evaluating half-life data.

1. Apparent loss caused by dilution resulting from plant growth.
2. Loss from the exterior surface. Suggested methods of loss are particle removal, volatilization, and cuticle flaking.
3. Loss from inside plants. There is very little translocation of iodine but iodine can possibly escape via transpiration or other gaseous exchanges.

The significance of these three routes has not been evaluated but must be understood in order to accurately predict effective half-lives. Experiments done in this lab show that the effective decay rate of iodine from plants occurs in two phases. At first the loss rate is rapid, presumably caused by the loss of surface contamination. Later the loss rate is less with dilution by growth and loss of absorbed iodine becoming the principal processes.

Various authors working with radioiodine have reported their results on the basis of wet weight, dry weight, leaf area, and the total ground area covered by plants. This paper presents evidence to show that areas of ground cover and wet weight are poor bases for data expression. Dry weight, although not perfect, is a much sounder basis for expressing contamination data than either of the others. Predictions of contamination cannot accurately be made until we understand the differences between species and these will never be understood unless there is a standard method for data expression.

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APPENDIX A. Deposition Velocity of ^{131}I.

Dep. Vel. V_g = cm/sec	Contaminant ^{131}I	Species	Density of cover gm/m^2	Wind m/sec	RH %	Temp °C	Reference	Remarks
0.01†	CH_3I	Grass	300[W]	15*	33	17.8	3	No ^{131}I was detected on samples
0.82	V	Grass		5.4*	34	9.3	3	
0.30		Grass					104	Windscale accident N. England
0.11		Grass					104	Windscale accident S. England
0.25		Grass					46	S.L. accident 1 KM from release
0.21		Grass					46	8.5 KM from releasee
0.23		Grass					46	67 KM from release
1.53 ± 0.59		Grass					39	Grass in trays
2.2		Grass					73	
2.8		Grass					50	
0.6 ± 0.22	V	Grass	46[d]	7.1*			49	Grass 13 cm high
0.55	V	Sece	129[d] ± 36				27	
0.59	V	Sece	65[d] ± 15				27	
0.52	V	Artr	129[d] + 19				27	
0.35		Brte	75[d] ± 14				27	
0.33		Dry soil					27	
1.91 ± 0.32	V	Grass	500[W]	4.20**	10		20	Sunny
2.65 ± 0.50	V	Grass	200[W]	3.72**	18		20	Cloudy

APPENDIX A. Deposition Velocity of ^{131}I. (Continued)

Dep. Vel. V_g = cm/sec	Contaminant ^{131}I	Species	Density of cover gm/m^2	Wind m/sec	RH %	Temp °C	Reference	Remarks
1.79 ± 0.21	V	Grass	260[w]	4.42**		20	20	Sunny
3.75 ± 0.19	V	Grass	420[w]	3.10**		21	20	Sunny
1.72 ± 0.27	V	Grass	420[w]	1.38**		16	20	Nearly dark
0.5	≥1μ	Grass		4.47***			40	60-70 cm tall
1.0	10μ	Grass		4.47***			40	60-70 cm tall
2.0	V	Grass		4.47***			40	60-70 cm tall
3.0	20μ	Grass		4.47***			40	60-70 cm tall
0.5	2.5μ	Grass		Stable***		101		Zinc sulfide crystals
1.0	V	Sece		Stable***		101		20.4 cm leaf blade (58% top, 29% bottom, 13% soil)
0.2	1μ	Artr		2.5***		57		Strong inversion
5.56 ± 2.80	1μ	Artr		5.68***		57		Fluorescent particles

† = calculated V_g
± = 1 standard deviation
V = vapor
Sece = *Secale cerceal* (Rye grass)
Artr = *Artemesia tridentata* (Big sagebrush)
Brte = *Bromus tectorum* (Cheat grass)
w = density based on vegetation wet weight
d = density based on vegetation dry weight
*=wind measured at 4 m
**= wind measured at 1 m
*** = height of measurement not indicated

APPENDIX B. Effective Half-life of ^{131}I.

T_{eff} Days	Contaminant ^{131}I	Species	Density of Vegetation g/m^2	Method of Expression	Reference	Remarks
3.5	V	Agde	46[d]	Wet weight	49	CERT 1 wind 7.1 m/sec at 4 m 21.1°C 5.2%/day = rate of grass growth - 13 cm high
5.1	V	Pasture grass		Area	13	CERT 2
6.5	V	Pasture grass	300	Area	13	9.3°C 34% RH, wind 5.4 m/sec CERT 7
5 to 6	V	Grass		Area	20	Based on five trials
5.0	V	Mixture grass & forbs		Area	76	Rain 65 mm on 18th day. No effect on T_{eff}
4.1 ± 0.3	V	Sece	129[d] ± 3.0	Area	27	
3.1 ± 0.5	V	Agde	6 ± 1.4	Area	27	Clipped 1.5 inches
4.0 ± 0.6	V	Artr	12 ± 1.8	Area	27	Leaves only
2.9 ± 0.3	V	Brte	7 ± 1.3	Area	27	
7.0 ± 0.3*	SD	Artr		Dry weight	112	Groom Valley 27 mi from GZ
8.4 ± 0.5*	SD	Atco		Dry weight	112	Penoyer Valley 44 mi from GZ
6.8 ± 0.4*	SD	Atco		Dry weight	112	Railroad Valley 70 mi from GZ
9.4 ± 0.8*	SD	Artr			112	Currant Valley 70 mi from GZ
4.9	V	Pasture			8	Windscale accident
5.0	V	Grass			8	

APPENDIX B. Effective Half-life of ^{131}I. (Continued)

T_{eff} Days	Contaminant ^{131}I	Species	Density of Vegetation g/m^2	Method of Expression	Reference	Remarks
5.8	WF	Chilton grass			89	Russian series in 1961
4.7	SD	Artr			70	Groom and Currant Valleys
5.5	SD	Av desert plants			70	All plants averaged--Groom, Currant, Penoyer, Railroad Valleys

± = 1 standard deviation
* = half-life was computed from data published by Turner & Martin[112] (see Appendix C)
V = vapor
SD = Sedan debris
WF = World-wide fallout
Agde = *Agropyron desertorum*
Sece = *Secale cerceal*
Artr = *Artemesia tridentata*
Brte = *Bromus tectorum*
Atco = *Atriplex confertifolia*
d = density based on dry weight
Area = ground covered by plant sample

APPENDIX C. Half-life of ^{131}I on Vegetation Contaminated with Sedan Fallout*.

Half-life was computed for samples collected at each point even though there was no replication in the sampling. Variation between points was so great that grouping of points was considered invalid. Half-lives for each area were computed by taking the mean of all points and compounding the errors.

C-1. Groom Valley, Nevada

Point	2	3	4	6	7	8
Time-Days			Activity (pCi/g of vegetation)			
5	103	22433	873	19369	8063	9082
10	125	14134	629	11622	351†	
15	60.5	5330	410	6089	2090	2040
20	101	3305	85.9	738	1605	389
25	542	1223	97.8	302	578	428
30	35.0	500	27.3	506	399	252
60	2.9	59.9	6.9	15.6	33.6	15.9
T_{eff}	9.3 ± 1.2	6.3 ± 0.6	7.5 ± 1.1	5.3 ± .7	7.1 ± .5	6.3 ± .8

Note--Mean half-life 7.0 ± .3
*Adapted from Turner and Martin, 1963(112)
†Omitted from analysis

APPENDIX C. Half-life of ^{131}I on Vegetation Contaminated with Sedan Fallout.*

C-2. Penoyer Valley, Nevada

Point	1	4	5	7	9	20
Time-Days			Activity (pCi/g of vegetation)			
5	3656	681.5	467	745.5	800	
10	1409	360		2299†	402.5	1301.5
15	989	260	265	189	104	567.5
20	466	124	60.9	111	152	196
25		56.8	54.6	144	198	196
31	219		40.0	102	50.0	197
61	31.8	4.9	3.6	7.1		12.9
T_{eff}	8.7 ± .8	8.0 ± .6	8.0 ± .9	9.0 ± 1.0	8.0 ± 2.4	8.4 ± 1.0

Note--Mean half-life 8.4 ± .5
†Omitted from analysis
*Adapted from Turner and Martin, 1963 (112)

APPENDIX C. Half-life of ^{131}I on Vegetation Contaminated with Sedan Fallout.*

C-3. Railroad Valley, Nevada

Point	2	3	4	6	7
Time-Days			Activity (pCi/g of vegetation)		
5	985		752	675	608
10	475.5	353	242.5	422	389
15	228	188	129	267	242
20	86.9	85.9	151		91
25	90.1	74.1	70.5	143	99.6
30		44.6	22.7	65.1	87.3
T_{eff}	5.3 ± .8	6.8 ± .8	5.8 ± .9	7.9 ± .6	8,2 ± 1.4

Note--Mean half-life 6.8 ± .4

*Adapted from Turner and Martin. 1963. (112)

APPENDIX C. Half-life of ^{131}I on Vegetation Contaminated with Sedan Fallout.*

C-4. Currant, Nevada

Point	2	3	4	5
Time-Days	Activity (pCi/g of vegetation)			
5	596	99.2	653	324
12	142	111	70	225
16	63	49	48	53
21	29	35	14	60
26	21	22.3	10.5	44.6
31	27.3	27.3	104	24.6
62	4.3	3.7	1.6	4.3
T_{eff}	9.1 ± 1.8	11.4 ± 1.4	7.6 ± 1.9	9.4 ± 1.2

Note--Mean half-life 9.4 ± .8
*Adapted from Turner and Martin 1963. (112)